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Plutonium Decontamination of Uranium using CO_2 Cleaning

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U.S. Department of Energy

Lawrence
Livermore
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Introduction

Purpose and Scope

A concern of the Department of Energy (DOE) Environmental Management (EM) and Defense Programs (DP), and of the Los Alamos National Laboratory (LANL) and the Lawrence Livermore National Laboratory (LLNL), is the disposition of thousands of legacy and recently generated plutonium (Pu)-contaminated, highly enriched uranium (HEU) parts. These parts take up needed vault space. This presents a serious problem for LLNL, as site limit could result in the stoppage of future weapons work. The Office of Fissile Materials Disposition (NN-60) will also face a similar problem as thousands of HEU parts will be created with the disassembly of site-return pits for plutonium recovery when the Pit Disassembly and Conversion Facility (PDCF) at the Savanna River Site (SRS) becomes operational.

To send HEU to the Oak Ridge National Laboratory and the Y-12 Plant for disposition, the contamination for metal must be less than 20 disintegrations per minute (dpm) of swipable transuranic per 100 cm^2 of surface area or the Pu bulk contamination for oxide must be less than 210 parts per billion (ppb). LANL has used the electrolytic process on Pu-contaminated HEU weapon parts with some success. However, this process requires that a different fixture be used for every configuration; each fixture cost approximately $10K. Moreover, electrolytic decontamination leaches the uranium metal substrate (no uranium or plutonium oxide) from the HEU part. The leaching rate at the uranium metal grain boundaries is higher than that of the grains and depends on the thickness of the uranium oxide layer. As the leaching liquid flows past the HEU part, it carries away plutonium oxide contamination and uranium oxide. The uneven uranium metal surface created by the leaching becomes a trap for plutonium oxide contamination.

In addition, other DOE sites have used CO_2 cleaning for Pu decontamination successfully. In the 1990's, the Idaho National Engineering Laboratory investigated this technology and showed that CO_2 pellet blasting (or CO_2 cleaning) reduced both fixed and smearable contamination on tools (Ref.1). In 1997, LLNL proved that even tritium contamination could be removed from a variety of different matrices using CO_2 cleaning (Ref. 2).

CO_2 cleaning is a non-toxic, nonconductive, nonabrasive decontamination process whose primary cleaning mechanisms are

- Impact of the CO_2 pellets loosens the bond between the contaminant and the substrate.
- CO_2 pellets shatter and sublimate into a gaseous state with large expansion (~800 times). The expanding CO_2 gas forms a layer between the contaminant and the substrate that acts as a spatula and peels off the contaminant.
- Cooling of the contaminant assists in breaking its bond with the substrate.

Thus, LLNL conducted feasibility testing to determine if CO_2 pellet blasting could remove Pu contamination (e.g., uranium oxide) from uranium metal without abrading the metal matrix. This report contains a summary of events and the results of this test.

LLNL CO_2 Cleaning Feasibility Study

On October 1, 2000, LLNL began a feasibility study to determine whether CO_2 pellet blasting could remove uranium oxide from uranium metal without abrading the metal matrix. The Nuclear Materials Focus Area (NMFA) and DOE-DP proposed $200K and $300K for work on this project, respectively. However, LLNL did not receive any funding from the NMFA by October 1.

On February 13, based on findings in an HEU report (Unallocated Off-Specification HEU: Recommendation for Disposition, in draft), the NMFA notified LLNL that it would only provide $50K for a feasibility paper study instead. By this time, however, LLNL was well into the study. We had taken full ownership of and developed new procedures for the CO_2 cleaning system in the Tritium Facility, removed existing equipment and tritium from the system glovebox, and started design work on test fixtures. Therefore, it made little sense at this point to stop the project.

In March, a representative from the Laboratory went to Washington, DC, to review the HEU report. Several concerns were identified, and as a result NMFA increased funding for the CO_2 cleaning project to $150K.

In April, the first test fixture was installed in the CO_2 cleaning glovebox and all room safety instruments were brought online. The stationary fixture is connected to a 2000-ft^3/min, high-efficiency particulate air (HEPA)-filtered vent line and uses a CO_2 nozzle that rotates on an axis perpendicular to the test part. Figures 1–3 show the test fixture inside the glovebox.

Figure 1. Test fixture in glovebox with front door opened.

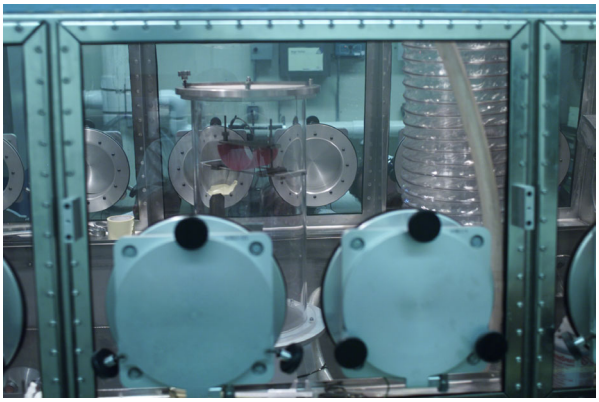


Figure 2. Test fixture in glovebox (side view).

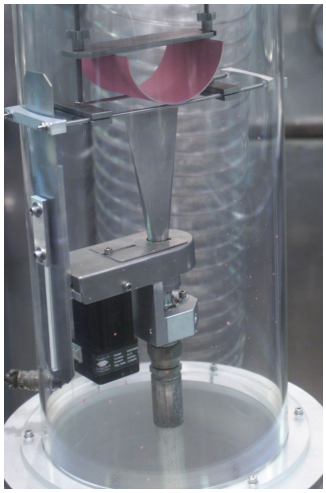


Figure 3. Test fixture ready for testing painted aluminum parts.

In May, the CO_2 pellet-making machine and the CO_2 pellet blaster were brought online. Blasting experiments were conducted using aluminum parts. The results from this series of tests showed that as pressure and time increased the abrasive power of the pellets also increased (see Figs. 4–8). This was most pronounced in the center of the part corresponding to the center of the blast nozzle.

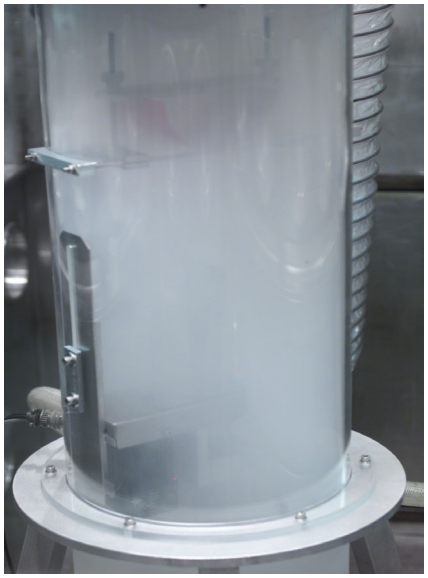


Figure 4. Test fixture testing painted aluminum part.

Figure 5. Painted aluminum part blasted at 45 psig for 15 sec.

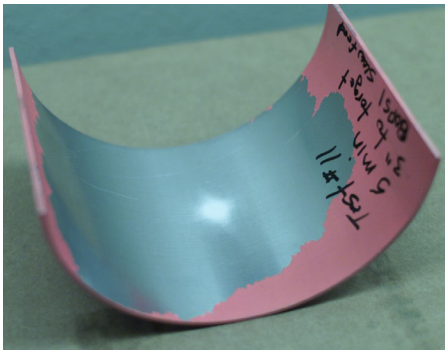


Figure 6. Painted aluminum part blasted at 80 psig for 5 min.

Figure 7. Anodized aluminum part blasted at 80 psig for 1 min.

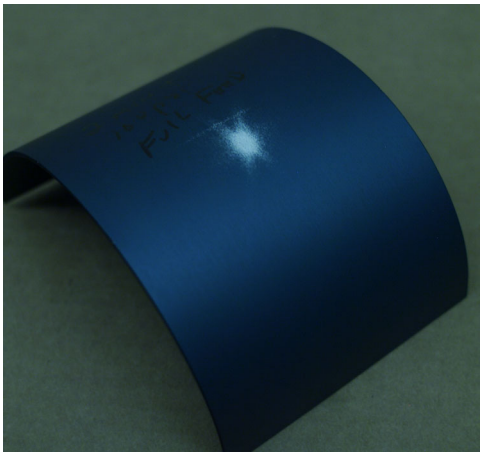


Figure 8. Anodized aluminum part blasted for at 100 psig for 3 min.

In June and July, cleaning experiments were conducted to determine if CO_2 cleaning could remove uranium oxide from depleted uranium metal without abrading the metal matrix. Using a surface electron microscope, we analysis the results and confirmed that this was possible. Moreover, the results showed that the cutoff for abrading the uranium matrix was at a pressure of 160 psig for 10 min with a nozzle distance of 0.5 in. (see Figs. 9–11). A comparison of Figures 9 and 10 shows that decreasing the nozzle distance from 2 in. to 0.5 in. exceeded the abrading cutoff point. However, major abrading occurred in a 0.25-in. circle centered within the

2-in. circle that was cleaned. Figures 11a and b show a small amount of abrading when the depleted uranium was blasted at 160 psig for 10 min with a nozzle distance of 0.5 in. (NOTE: The sample in this case was 0.5 in. from the center of 2-in. blast zone.

An element of surprise from these experiments was that all swipes taken inside the test fixture showed no alpha contamination. This suggests that recontamination would not be a problem using the CO_2 cleaning method.

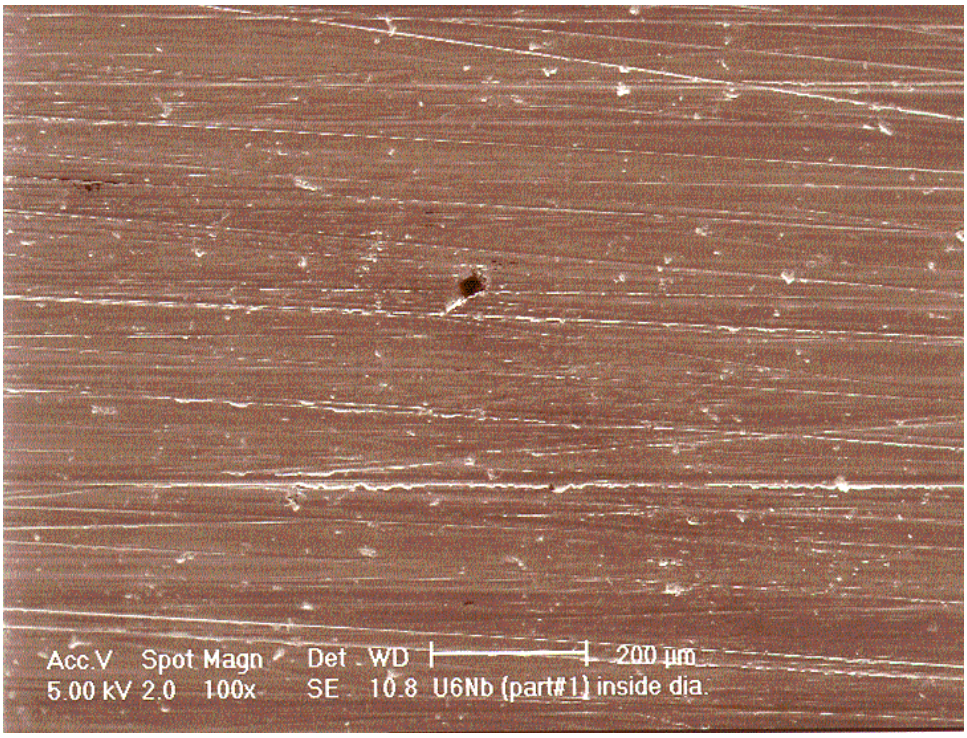


Figure 9a. Depleted uranium blasted at 160 psig for 10 min with a nozzle distance of 2 in. (NOTE: Sample was in the center of 2-in. blast zone).

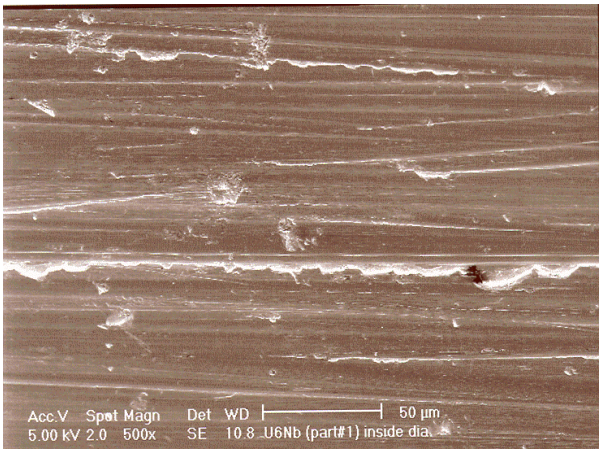


Figure 9b. Depleted uranium Blasted at 160 psig for 10 min with a nozzle distance of 2 in. (NOTE: The sample was in center of 2-in. blast zone).

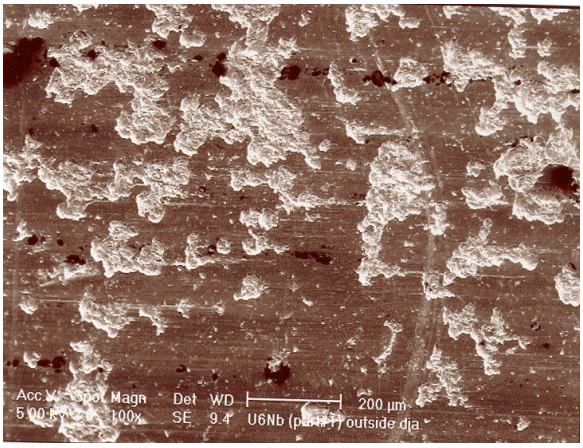


Figure 10a. Depleted uranium blasted at 160 psig for 10 min with a nozzle distance of 0.5 in. (NOTE: The sample was in center of 2-in. blast zone).

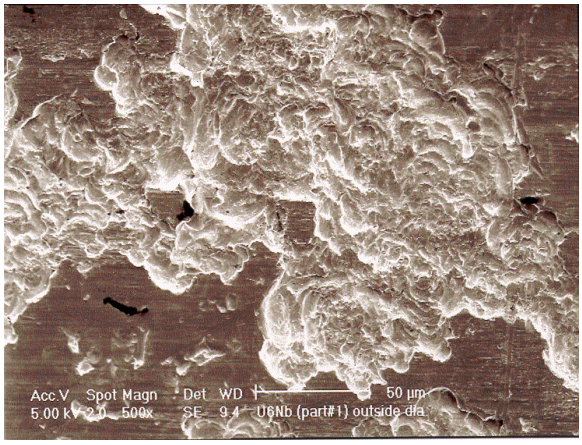


Figure 10b. Depleted uranium at 160 psig for 10 min with a nozzle distance of 0.5 in. (NOTE: The sample was in the center of 2-in. blast zone.)

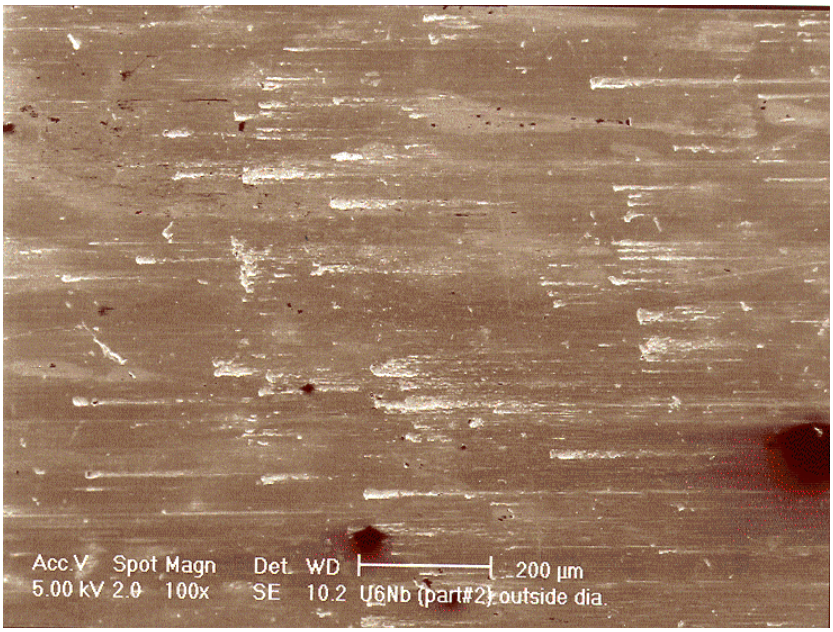


Figure 11a. Depleted uranium blasted at 160 psig for 10 min with a nozzle distance of 0.5 in. (NOTE: The sample was 0.5 in. from the center of 2-in. blast zone.)

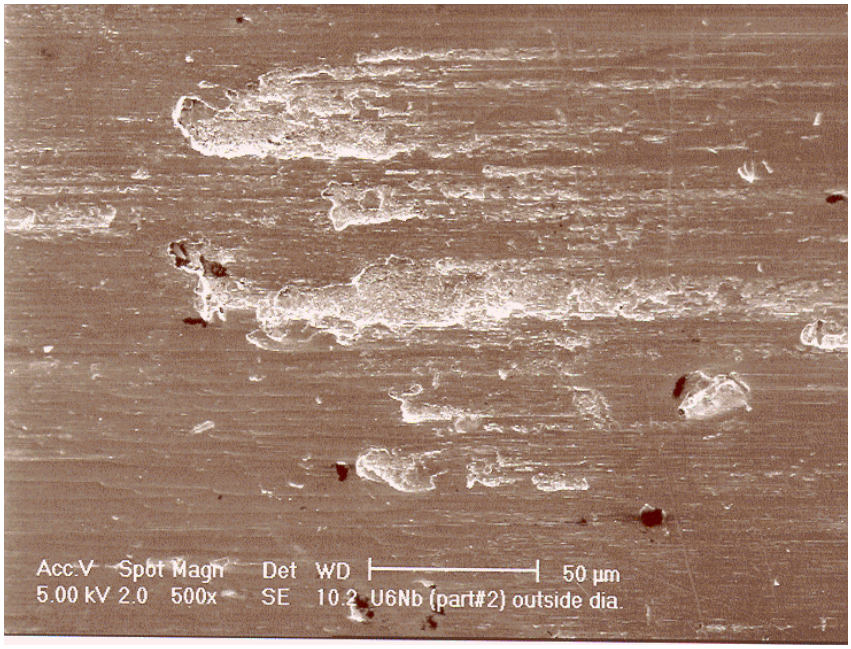


Figure 11b. Depleted uranium blasted at 160 psig for 10 min with a nozzle distance of 0.5 in. (NOTE: The sample was 0.5 in. from the center of 2-in. blast zone.)

Conclusion

The results of the feasibility test showed a high probability for developing a CO_2 cleaning method for decontaminating Pu-contaminated HEU shells to meet the Y-12 acceptance requirements for HEU metal. Moreover, there is even a higher probability that if the CO_2 cleaning process were used following HEU oxidation, the uranium oxide produced would meet the Y-12 Plant requirement for bulk HEU oxide. Thus, LLNL has designed, procured, and begun installation of a new fixture to test Pu-contaminated HEU samples (see Figs. 12a, b, and c). The new fixture will be tested using 1-in.-square coupons cut from Pu-contaminated uranium shells from LLNL inventory. If successful, LLNL will then demonstrate the CO_2 cleaning method using Pu-contaminated HEU shells.

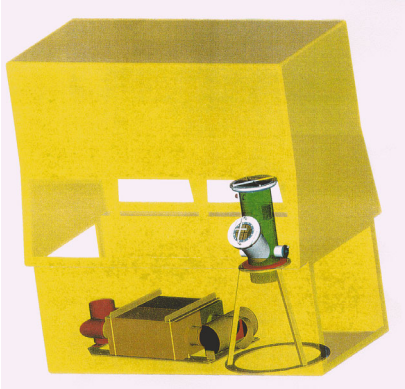
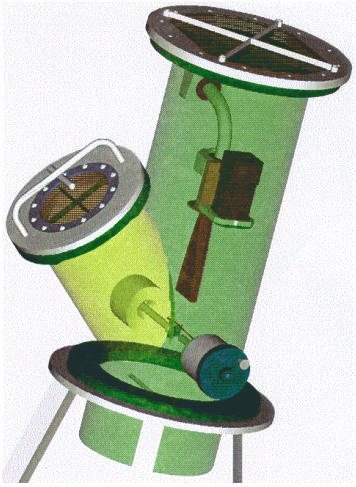


Figure 12a. New fixture in the Tritium Facility glovebox, with the CO_2 cleaning head on the right and two high-efficiency particulate air (HEPA) filters in parallel on the left. (NOTE: The ventilation lines between the fixture head and the HEPA filters are not shown).



Figures 12b. CO_2 cleaning test fixture head.

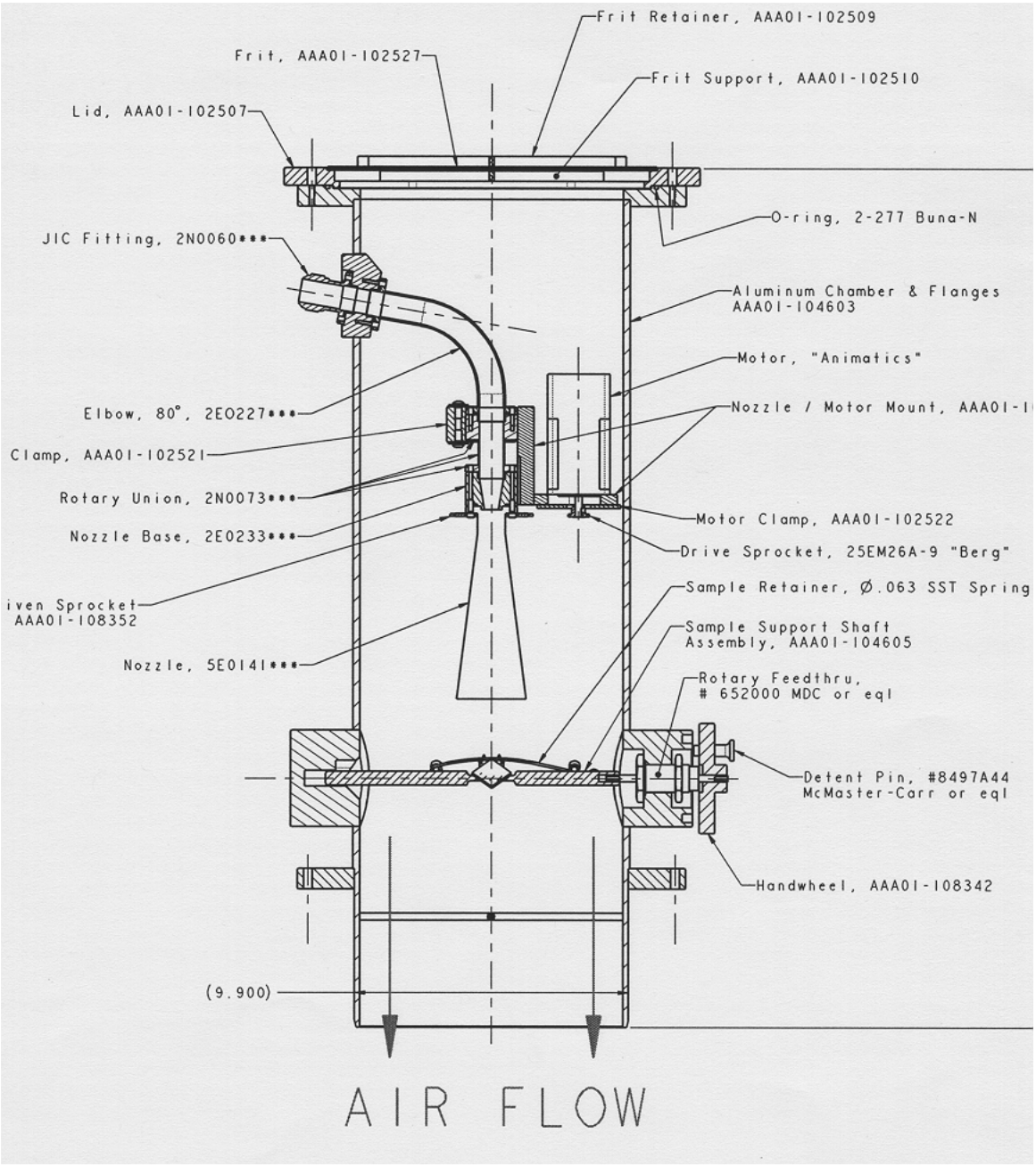


Figure 12c. Schematic of the CO2 cleaning test fixture head.

Acknowledgements

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References

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2. Memo from Roger M. Watson to Thomas Reitz and J. M. Mintz, Re: Co_2 Cleaning Station Decontamination of Secondary Containers, October 13, 1997 (TF07-076).

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